Application of Flexible Recipes for Model Building, Batch Process Optimization and Control

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Unlike the traditionally fixed recipes in batch process operation, flexible recipes allow the adjustment of some of its relevant recipe items. These adjustments can either be predefined in cases of planned experimentation, or suggested by a formal process optimization or control algorithm on the basis of actual information. In both the response surface methodology and the simplex evolutionary operation (EVOP), some well-known methods for empirical model building and process optimization, flexible recipes are involved. Another application of flexible recipes arises in a feedforward quality control strategy of batch processes when variations in market or process conditions are known a priori. The experimental results of these strategies are presented for the batchwise production of benzylalcohol on a pilot-plant scale. Experiments have been performed to obtain a reliable model of the yield. On the basis of this model, better process conditions have been suggested, which substantially deviate from the final simplex resulted from experiments within simplex EVOP. Finally, an adaptive feedforward control strategy has been applied for a priori known disturbances in the process inputs.

Introduction

Flexibility is one of the keywords in industrial activities today. Due to variations in both market and process conditions the plant operation must be adjusted in order to meet the specific final conditions of the product in terms of quality and quantity as well as to produce efficiently. Examples of market variations are changes in costs of raw materials and energy, selling-prices of the final products and product requirements. Apart from this, fluctuations in feed quality or quantity, fouling of the process equipment and temperature limitations will cause variations in the process operation conditions.

Since the late 70s it has been recognized that already in the *design* stage attention has to be paid to the flexibility of chemical processes for variations which might occur in the future. This flexibility was analyzed by incorporating uncertainty in the process parameters and constraints that appeared in the optimal design procedure (see, for instance, Grossman et al., 1983, for an overview; Grossmann and Floudas, 1987).

In this article, we will not focus on the design, but on the operation of flexible chemical processes. Note that, unlike the procedures for process design, we have to deal now with actual variations in market and process conditions. In our approach, flexibility in operation is obtained by flexible recipes which allow adjustments of some recipe items. The key idea behind this flexible recipe approach is that by deviating from the traditionally fixed recipes better performance can be obtained under a wide range of changing conditions. Usually, a recipe is defined as a set of predetermined instructions for the manufacturing of a specific product. In the flexible recipe context, however, the term recipe is used in a more abstract way by referring to a selected set of adjustable recipe items which control the process output. In the following, we will restrict ourselves to batch processes, or more specifically, to the reaction phase in a single batch unit under changing process conditions. In our context, these changes may also include variations imposed by an experimenter. It should be noted that, although batch processes exhibit more flexibility than continuous ones, the approach is certainly not restricted to batch processes.

Within the setting of evolutionary operation (EVOP) (see, for instance, Box, 1957; Box and Draper, 1969) flexible recipes have been introduced in process operation for the first time.

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Their main objective was to gain insight into the process behavior in a statistically elementary way in order to gradually improve the process efficiency. A more sophisticated approach and, in fact, an extension of EVOP, is the response surface methodology (RSM) (see Box and Draper, 1987) which involves more complicated experiment designs (inclusive possible transformations of the process variables and responses) as well as a formal analysis of the response surface. A well-known alternative to this statistical approach, which is also based on the application of flexible recipes, is the so-called simplex EVOP (Spendley et al., 1962). In simplex EVOP, unlike the original EVOP, optimal process conditions are found empirically. These applications of flexible recipes have in common that the adjustment of the recipe is performed within a feedback loop which includes one or more batch cycles.

Recently, a feedforward (quality) control strategy has also been proposed (see Keesman, 1991; Rijnsdorp, 1991). In this strategy, the adjustments of the relevant recipe items are calculated from a (non)linear program, assuming that the variations in market and process conditions are known *a priori*. This application of flexible recipes is indicated as *recipe initialization*. Apart from recipe initialization, the recipe can also be adjusted during the batch cycle when in-line measurements of the process conditions become available. However, these recipe adjustments within a feedback loop during a batch cycle is out of the scope of the article.

The purpose of this article is to present a real-world, industrial application of flexible recipes for empirical model building, process optimization, and quality control. The article has been organized as follows. The second section is devoted to the region of applicability for both RSM and simplex EVOP, and some essential tools within these procedures are discussed. Furthermore, an adaptive recipe initialization strategy is proposed which accounts for structural changes in process operation. The third section gives the results of the application of flexible recipes for a batchwise production of benzylalcohol. Since all these applications have, in essence, an iterative character the evaluation of information as a function of the experimental run number will be emphasized in the discussion of the results.

Methods

The key ideas behind empirical model building have been published extensively (see, for instance, Box and Draper, 1987). Therefore, only some crucial steps in this "black-box" modeling process are shortly outlined.

First, an appropriate model structure must be chosen. Commonly, when no dynamics are involved and the experimental region is limited, one starts with a simple first-order polynomial approximation of the underlying process. For a single variable system the model, assuming small deviations in the nominal process conditions and additive uncertainty, can be represented as:

$$\delta y(\beta,\delta x) = \delta x^T \beta \tag{1a}$$

$$z = \beta_0 + \delta y(\beta,\delta x) + e \tag{1b}$$

where δy is the variation in model response and β_0 is the nominal model response. Furthermore, the vectors δx, $\beta(\in R^k)$ contain, respectively, (coded/transformed) variations in the selected flexible recipe items, which are the inputs of the model, and model parameters. The observed process output and associated error are represented by z and e.

Secondly, to efficiently estimate the (unknown) model parameters from the data, an approximate experiment design must be specified. A well-known design for this class of models is the 2^k factorial design in which each input occurs at just two levels. Observe, then, that in this model building stage the experimenter-imposed variations on the inputs can be interpreted in terms of flexible recipe manipulation.

Thirdly, the model parameters $(\beta_0,\beta_1,\ldots,\beta_k)$ can be estimated from the experimental data set $\{X,z\}$, where $z \in R^N$ and the $N \times (k+1)$ matrix X includes the column vector $\mathbf{1}$. A very popular estimation procedure for this kind of model and data is the least-squares method which, under the assumption that the residuals are statistically independent with constant variance and are normally distributed, offers maximum likelihood estimates. These conditions allow an exact interpretation of the uncertainty in the parameter estimates of the linear model. However, when the number of measurements is small there is no detailed uncertainty structure characterization in terms of independency and normality. Moreover, hypotheses with respect to a detailed uncertainty structure cannot be tested. Recently, as an alternative to this stochastic approach, a so-called set-membership approach has been proposed for parameter estimation from small data sets (see Walter and Piet-Lahanier, 1990, for a recent survey). In this approach, the only assumption with respect to the uncertainty is that it is point-wise bounded. Hence, a set of feasible parameter vectors instead of a single optimal parameter vector will be found. Obviously less detailed information about the parameter estimate uncertainty, in terms of parameter bounds or individual feasible realizations (Keesman, 1990), is offered. Apart from the feasible set estimation, several different algorithms can be chosen if a specific point estimate is desired as, for instance, the central estimate (Milanese and Tempo, 1985) or the maximally robust (min-max) estimate (see Tempo et al., 1988; Keesman and van Straten, 1989). Whensoever reliable model parameter estimates are available, a response surface analysis can be performed in order to gain insight into the relation between response and inputs (Eq. 1a).

Finally, the set of residuals must be analyzed to verify the presuppositions made with respect to the uncertainty structure. When it appears that, for instance, the residuals are not serially uncorrelated or the maximum of the min-max residuals, which has not been detected as an outlier, is too large, the model structure must be modified in terms of increasing model order or variable transformation. Obviously, this modification step implies a new iteration in the modeling process.

On the basis of the response surface associated with the finally accepted model, a new set of recipe item values, which will most likely provide a higher yield, can be specified. As Box (1957) already pointed out, EVOP and in its more sophisticated form RSM basically rely on "scientific" instead of "empirical" feedback. If, however, the response surface changes as a result of process disturbances that are difficult to specify such as, for instance, fouling of the process equipment or catalyst deactivation, empirical feedback would be preferred. An effective empirical optimization technique, which uses a simplex instead of a factorial design, has been proposed

by Spendley et al. (1962). Nelder and Mead (1965) modified this method for function minimization. For application of the Nelder-Mead method in a bounded input space, Verwater (1990) proposes an additional operation, namely, shadow contraction.

So far, recipe adjustments for process optimization have been considered, and a distinction was made between model-based and empirical optimization. However, in both cases recipe adjustments occur on the basis of measured deviations in the process output. The question that arises then is: what happens when this deviation is large? As a result of replication of the design, large random deviations are filtered out in RSM. In simplex EVOP, on the contrary, the evaluation of the vertices in a simplex will be affected directly as will be demonstrated in the application.

Now, in addition to these more or less conventional methods, a novel adaptive feedforward control strategy for process quality control will be proposed, which will allow us to accommodate the process conditions to *a priori* known variations. Consider, therefore, the block diagram of Figure 1 in which there are only external process disturbances. Clearly, the disturbances (d) must be elements of the input vector $\delta x \in \mathbf{R}^k$ to calculate the compensation on the basis of the model (Eq. 1a), so that $\delta x = (d_1,\ldots,d_m,\delta u_1,\ldots,\delta u_{k-m})^T$ where u_i is a control input variable. Let the disturbances d and the model parameter estimates b be known, and let us try to maximize the yield y. The bounded control variables can then be calculated from the linear program (LP),

$$\max_{\delta u} \{\delta y(\boldsymbol{b},\boldsymbol{d},\delta \boldsymbol{u})\} \qquad |\delta u_i| \le v_i \;\; \forall i = 1,\ldots,k-m \qquad (2)$$

Note that the estimate b_0, the average process output for $\delta x = \mathbf{0}$, can be omitted in this LP problem, and that linear quality constraints on δy can easily be added (Keesman, 1991). The bounds on δu result from operability and model validity considerations. After implementation of the resulting values in the recipe, the reaction can be performed under the adjusted process operation conditions. At the end of the reaction phase, when the process output measurement becomes available, the parameters should be updated (see Figure 1) in order to keep the model as close as possible to the real process. The model parameters are adapted using a recursive least-squares algorithm with a random walk model for the parameters (see, for instance, Young, 1984). The incorporation of the random walk model,

$$\beta_j = \beta_{j-1} + q_{j-1} \qquad (3)$$

where q_{j-1} is a $k \times 1$ white noise vector of serially-independent random variables with zero mean and covariance matrix Q at run number $j-1$, allows a rapid change in the parameters, so that the model can be adjusted instantaneously. The problem, however, is the choice of Q which is a tradeoff between tracking ability and noise sensitivity.

The linear model will become invalid when the process is operated near a (local) unconstrained optimum or when the operability region is largely extended. Under these circumstances, a second-order model (possibly with transformed variables) will sufficiently describe the response in most applications, that is

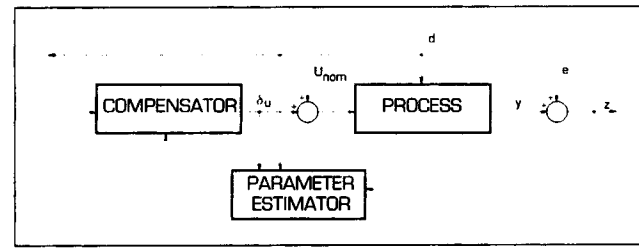

Figure 1. Feedforward control system for batch processes.

$$\delta y(\beta,\mathbf{B},\delta x) = \delta x^T \beta + \delta x^T \mathbf{B} \delta x \qquad (4)$$

where $\mathbf{B}$ is a symmetric square $(k \times k)$ matrix. The LP problem of Eq. 2 transforms then into a quadratic programming problem. Moreover, when a bound on the 2-norm of $\delta x (\|\delta x\|_2)$ is introduced, a nonlinear programming (NLP) problem results.

Application

In this section, we present the results of RSM for model-building, simplex EVOP (with a modification to the Nelder-Mead method) for process optimization, and the adaptive feedforward control strategy for the control of the final product quantity. These flexible recipe applications have been tested on the production of benzylalcohol in a pilot plant. The reaction equation for the reduction of benzaldehyde to benzylalcohol is:

benzaldehyde + formaldehyde + potassium-hydroxide → benzylalcohol + potassium-formate

which is an example of a so-called crossed-Cannizarro reaction (see Figure 2).

For our application four input variables were selected: temperature of reaction, time of reaction, amount of potassium-hydroxide (KOH), and amount of formaldehyde. The measured process output is the conversion (%) from benzaldehyde to benzylalcohol, which has been analyzed in a Gas-Liquid Chromatography (GLC) device.

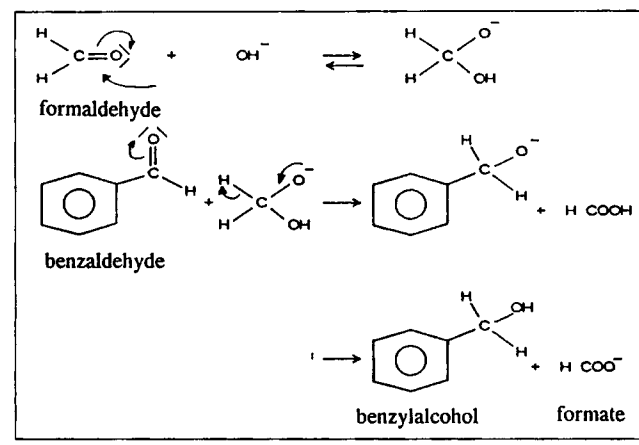

Figure 2. Reaction mechanism.

Table 1. Experimental Region

Coded Levels	δx_i	−1	+1	
Reaction Temperature	ξ_1	63	65	(°C)
Reaction Time	ξ_2	1	2	(h)
Amount of KOH	ξ_3	450	550	(g)
Amount of Formaldehyde	ξ_4	375	475	(g)

Model-building

In the modeling stage, experiments were performed on the basis of a two-level design for four inputs with center points. The experimental region is presented in Table 1, where

$$\delta x_1 = (\xi_1 - 64)/1, \quad \delta x_2 = (\xi_2 - 1.5)/0.5,$$

$$\delta x_3 = (\xi_3 - 500)/50, \quad \delta x_4 = (\xi_4 - 425)/50.$$

To reduce the effect of systematic disturbances on main effects and the significant interactions, the experiments were arranged in blocks. When only one half of the total number of blocks is run, we speak about a 2^{4-1} fractional factorial design. From the first 10 experiments, which constitute the so-called first half-fraction of the 2^4 factorial plus two center points, the five parameters of the linear model (Eq. 1a) can efficiently be estimated (see Table 2 for the results). In addition to the parameter estimates the associated standard deviation (s_b), for which an approximated value for all parameters is represented, and the standard deviation of the residuals (s) are also presented in the table. However, for this small data set, the assumptions for an unbiased estimate from the ordinary least-squares method are questionable. Therefore, in addition to the least-squares estimate, the min-max estimate for bounded errors (that is, $\|e\|_\infty := \max_{j=1,\ldots,N} |e_j| \le \epsilon$ is also added in Table 2. In this linear case, the min-max estimate can easily be found by solving the following LP problem with $k+2$ parameters and $2N+1$ constraints (Dem'yanov and Malozemov, 1974),

$$\min_{\epsilon, b_0 \in R, b \in R^k} \{\epsilon\} \quad \epsilon \ge 0 \quad z_j - \epsilon \le b_0 + \delta x_j^T b \le z_j + \epsilon, \; j = 1, \ldots, N \tag{5}$$

After running the second half-fraction plus two center points one cycle has been completed. From this cycle, in addition to the linear parameters, the two-factor interaction parameters (denoted as b_{ij}) can also be estimated efficiently.

The addition of center points to the 2^4-design allows us to test for second-order curvature, since the expectation of the contrast between the average response at the factorial points ($\bar{z}$) and the average response at the center ($\bar{z}_0$) is equal to the trace of matrix **B**, that is

$$E[\bar{z} - \bar{z}_0] = \sum_{i=1}^{k} \mathbf{B}_{ii} \tag{6}$$

(see Box and Draper, 1982). It appears that this contrast is equal to +0.5 for the first cycle. Note that a small value does not necessarily mean that there is no curvature of the response surface; the parameters $\mathbf{B}_{ii}$ can be of the opposite sign.

To separately quantify the pure error, consisting of setup errors, sampling errors and analytical errors, and the lack of fit due to incompleteness of the model, the cycle has to be repeated at least once. Moreover, additional experiments will also contribute to more reliable parameter estimates. The parameter estimates, the mean-squared errors associated with the pure error (MS_e) and the lack of fit for the linear model with interaction terms (MS_L) after two cycles are presented in the eighth column of Table 2. In addition to the separate mean-squared errors the rate $F_L = MS_L/MS_e$, a statistic which allows a formal hypothesis testing of the model validity, it also represented. It can easily be verified that the diagonal elements

Table 2. Model-Building Results

Runs	10		20		40		59			
	LS*	MM	LS	MM	LS	MM	LS	MM	LS	MM
b_0	67.0	67.6	68.0	67.9	67.8	68.2	67.5	67.7	67.7	70.8
b_1	4.9	4.0	4.6	4.4	4.5	4.7	4.5	4.4	4.4	5.6
b_2	5.4	8.0	6.7	5.4	6.7	6.5	7.1	6.8	8.1	6.9
b_3	1.8	0.9	2.8	1.3	3.1	2.8	2.9	3.0	1.9	1.6
b_4	3.0	5.7	2.3	3.0	2.0	2.1	2.7	3.0	1.9	2.3
b_{12}**					−2.1	−2.0	−1.5	−1.0	−1.3	0.7
b_{13}					−2.0	−1.6	−2.3	−2.5	−2.3	−3.6
b_{14}					1.8	1.1	2.3	2.2	2.3	3.1
b_{23}					1.1	1.1	1.4	1.2	1.4	3.6
b_{24}					1.3	1.6	0.8	0.4	0.8	0.5
b_{34}					−3.2	−3.0	−3.1	−2.7	−3.1	−3.8
b_{11}									0.5	−0.7
b_{22}									−2.9	−3.4
b_{33}									3.3	3.2
b_{44}									−0.4	−0.9
s_b†	2.5		1.4		0.9		0.5		0.6	
s	7.4		5.5		2.5		2.7		3.7	
ϵ		7.8		7.9		2.4		3.5		6.6
MS_L							22.6		41.0	
MS_e							3.2		5.6	
F_L							7.0		7.3	

*LS = least-squares estimate; MM = min-max estimate.
**Note that $b_{ij} = 2B_{ij}$ for $i \neq j$, and $b_{ii} = B_{ii}$ (see Eqs. 4 and 7).
†Approximate value for all estimates.

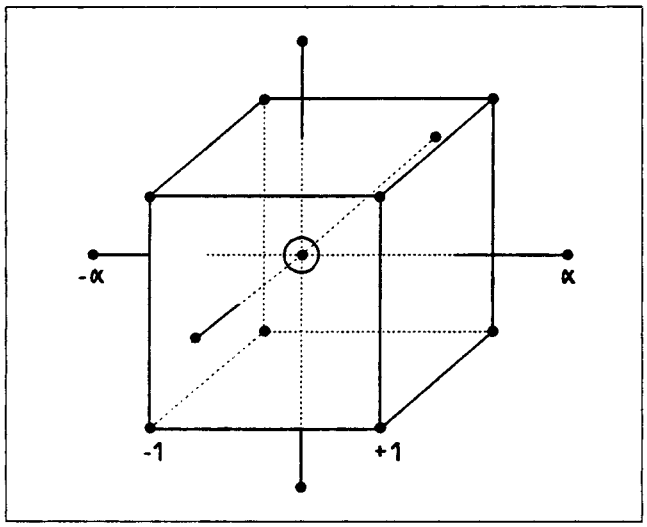

Figure 3. Composite design (cube, star and center points) for $k=3$.

of **B**, unlike the off-diagonal elements, cannot be estimated efficiently from our experiments based on the first-order design with center points.

For quadratic models (Eq. 4) Box and Draper (1987) recommend a second-order composite design (see Figure 3) consisting of: a "cube" and "star" part of the design with center points. The "cube" part compromises points with coordinates of the type $(\pm 1, \pm 1, \ldots, \pm 1)$. The axial points with coordinates $(\pm\alpha,0,\ldots,0), (0,\pm\alpha,\ldots,0),\ldots,(0,0,\ldots,\pm\alpha)$ form the "star" part of the design. And, the point $(0,0,\ldots,0)$ is the center point. In Table 15.2 of their book, useful second-order composite designs for different values of k are presented. In order to obtain a rotatable design α must be equal to 2 for $k=4$. In addition to the 40 runs from the first-order design another two cycles of 10 runs each have been performed on the basis of the "star" design in order to complete the composite design. From a first analysis, it appeared that one run was invalid, so that the parameters of our second-order model had to be estimated from the remaining 59 experiments. The effect of removing this outlier on parameter b_{12} can be seen in Table 2. Note also that the parameters b_{24}, b_{11} and b_{44} could be considered as nonsignificant. However, we have chosen to keep these parameters in the model, since removal of these terms hardly affected the other estimates, and it kept the model more flexible for adaptation to changing conditions. The resulting second-order model is, then,

$$\begin{aligned}
\delta y = {} & 4.4\ \delta x_1 + \underset{(\pm 0.6)}{8.1\ \delta x_2} + \underset{(\pm 0.6)}{1.9\ \delta x_3} + \underset{(\pm 0.6)}{1.9\ \delta x_4} \\
& - \underset{(\pm 0.7)}{1.3\ \delta x_1 \delta x_2} - \underset{(\pm 0.7)}{2.3\ \delta x_1 \delta x_3} + \underset{(\pm 0.7)}{2.3\ \delta x_1 \delta x_4} \\
& + \underset{(\pm 0.7)}{1.4\ \delta x_2 \delta x_3} + \underset{(\pm 0.7)}{\mathbf{0.8}\ \delta x_2 \delta x_4} - \underset{(\pm 0.7)}{3.1\ \delta x_3 \delta x_4} \\
& + \underset{(\pm 0.5)}{\mathbf{0.5}\ \delta x_1^2} - \underset{(\pm 0.5)}{2.9\ \delta x_2^2} + \underset{(\pm 0.5)}{3.3\ \delta x_3^2} - \underset{(\pm 0.5)}{\mathbf{0.4}\ \delta x_4^2} \qquad (7)
\end{aligned}$$

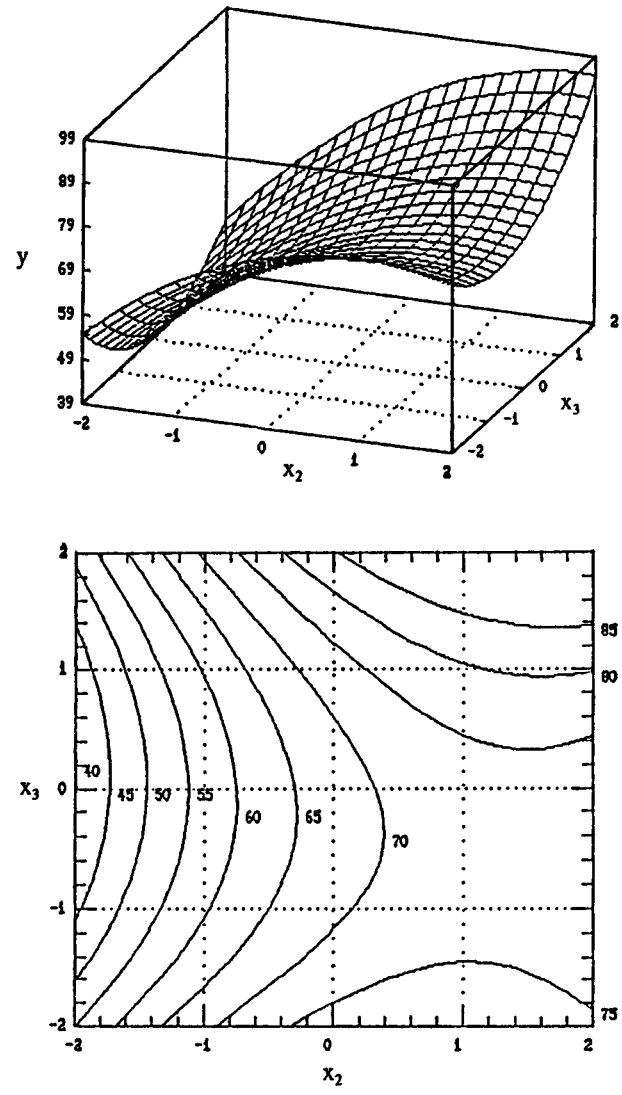

Figure 4. Response surface and contour lines for reaction time and amount of KOH.

and b_0 is 67.7($\pm$1.1), where the number between the parentheses are the standard deviations of the parameter estimates.

In Figure 4 the response surface and the associated contour lines for x_2 (time of reaction) and x_3 (amount of KOH) are presented. Notice that the response surface in this figure is a minimax, which will obstruct an empirical process optimization procedure. Similar surfaces have been obtained for x_1-x_2, x_1-x_4, x_3-x_4 with $|\delta x_i| \le 2$ for $i=1,\ldots,k$.

In fact, we can perform now a formal optimization on the basis of our model (Eq. 7), which lead to the optimum $\delta x_{opt} = (-0.19, 0.72, 1.82, -0.37)^T$ with $\|\delta x_{opt}\|_2 = 2$ and a conversion of 89.9%. Notice that the optimum is situated on the border of the experimental region. However, in practice, such an approach is hardly allowed in view of the uncertainty in the response surfaces. Under these circumstances, a more acceptable approach is to investigate the graphical representations of the response surface. Obviously, this can only be realized when one or two variables are involved. For studies with more than two variables a canonical analysis of the model can be performed (see Box and Draper, 1987). In this analysis,

Table 3. Starting Simplex

Vertex		1	2	3	4	5	
Reaction Temperature	ξ_1	63	65	64	64	64	(°C)
Reaction Time	ξ_2	1.16	1.16	2.16	1.5	1.5	(h)
Amount of KOH	ξ_3	475	475	475	575	500	(g)
Amount of Formaldehyde	ξ_4	405	405	405	405	505	(g)

the coordinate axes system is rotated in order to remove the interaction terms, which allows an easier interpretation of the response surface. Our second-order model transforms then into,

$$\delta y(\theta,\Lambda,\delta x') = \delta x'^T\theta + \delta x'^T\Lambda\delta x' \tag{8}$$

where $\delta x' = M^T\delta x$, and $\theta = M^T\beta = (-0.1,\ -4.9,\ -3.1,\ 7.6)^T$, in which M is a k × k matrix containing the eigenvectors of B. The diagonal matrix $\Lambda = \text{diag}(4.5,\ 0.3,\ -1.0,\ -3.3)$ contains the associated eigenvalues. In addition to the interaction terms, the linear terms can be removed from the model by shifting the axes system to the stationary point in the rotated system, $\delta x_s' = (0.0,\ 8.8,\ -1.6,\ 1.2)^T$, so that

$$\begin{aligned}\delta y(\Lambda,\delta x'') &= \delta y_s + \delta x''^T\Lambda\delta x'' \\ &= -14.6 + 4.5(\delta x_1' - 0.0)^2 + 0.3(\delta x_2' - 8.8)^2 \\ &\quad -(\delta x_3' + 1.6)^2 - 3.3(\delta x_4' - 1.2)^2 \end{aligned} \tag{9}$$

where δy_s is the deviation in the response at the stationary point $\delta x_s'$ and $\delta x'' = \delta x' - \delta x_s'$.

Notice that the direction of $\delta x_1'$ is dominant (λ_1 is largest eigenvalue). Let us select new process operation conditions on the basis of this information. For $\delta x' = (-2,\ 0,\ 0,\ 0)^T$, for instance, which is equivalent with $\delta x = (-0.7,\ 0.2,\ 1.7,\ -0.7)^T$, the yield is then 87.7%. It appears that this new point is rather close to the optimum which resulted from the preceding formal optimization. It should be noted that this approach, unlike the steepest ascent procedure, is essentially based on evaluation of the Hessian instead of the Jacobian of the model.

Process optimization

In the process optimization stage better operation conditions have been found by performing experiments which were based on a simplex design. The starting simplex for this procedure, enclosing the center point of Table 1, is presented in Table 3. For a comparison between these results and those from RSM, constraints have been imposed on the (coded) variables, that is, $\|\delta x\|_2 \le 2$. The evaluation of the simplices has been based on the Nelder-Mead procedure with a minor modification for the presence of constraints. In order to reduce the effects of analytical errors, which can disturb the search path significantly, each sample of the final product has been analyzed twice during the preparation phase of the next run.

The evaluation of the individual input variables δx_i in terms of their combined contribution to the squared norm, can be seen from Figure 5, where the first five bars are related to the starting simplex of Table 3. Note that the squared norm is smaller than or equal to four, which directly results from our restriction of the experimental region (a sphere with diameter two).

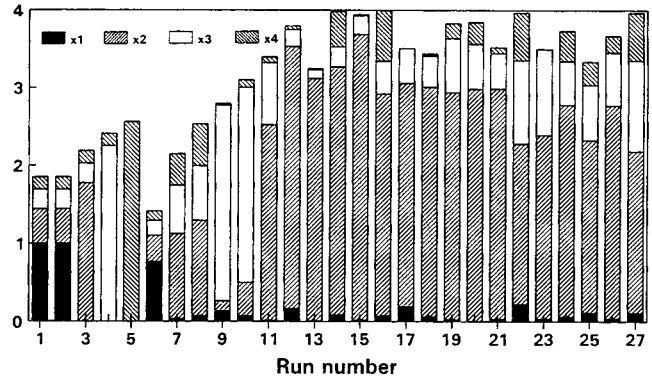

Figure 5. Evaluation of reaction temperature, reaction time, amount of KOH and formaldehyde in terms of δx_i^2.

From this graphical analysis, it appears again that in the selected experimental region both the reaction time (x_2) and the amount of KOH (x_3) are dominant in controlling the process output. However, unlike the results presented in the preceding section, the effect of the reaction time appears to be larger than the effect of KOH. Note from Figure 5 that in the tenth run process operation conditions that are close to the optimal conditions obtained from the preceding model-based optimization have been realized. The associated process output (73.4%) is, however, significantly lower than the predicted output for these conditions (85.5%) using the model of Eq. 7. In order to analyze this effect more specifically, the deviations between measured process output and predicted model response are presented for eighty-six experiments (see Figure 6).

The nonstationary conditions in the residuals coincides clearly with the start of the new experiment cycle at run number sixty. The disturbance is most likely the result of carryover effects; between the two types of experiments other experiments have been performed in the reactor.

The process improvement can most clearly be visualized by the measured process output as a function of the run number (see Figure 7). In (Nelder-Mead) simplex EVOP, the "optimal" process operation conditions are enclosed by the final simplex. The final simplex for our application is presented in Table 4.

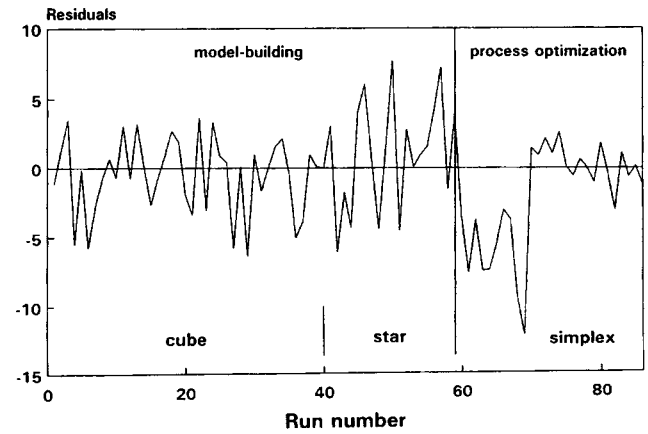

Figure 6. Model validation test.

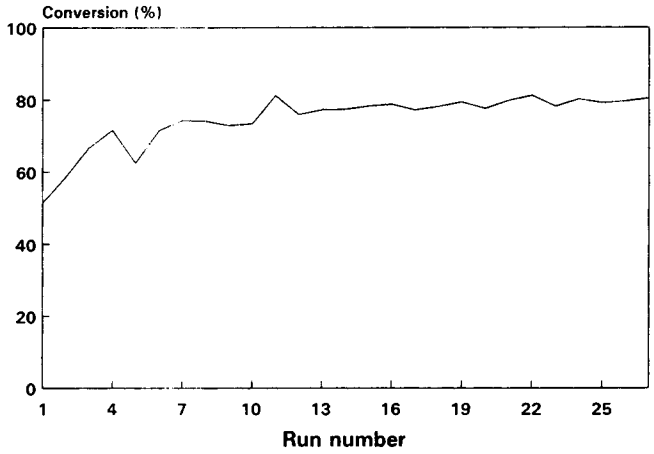

Figure 7. Measured process outputs in the process optimization stage.

Note the relative small variations in reaction time ($\delta x_2 = 0.22$) and amount of KOH ($\delta x_3 = 0.32$) which would support our conclusion that these variables are dominant if all variables were independent. For a more complex interpretation of the dominance of (a linear combination of) variables, we should perform an eigenvalue decomposition analysis of the matrix B. From the preceding section, in which this analysis has been performed, we know that the response surface is a minimax, so that a different simplex may have been resulted when, for instance, the starting simplex had been mirrored with respect to the origin. This fact clearly shows the problem one can run into when implementing an empirical process optimization procedure without evaluating the response surface.

Adaptive feedforward control

In this section, the experiment results of the adaptive feedforward control strategy with respect to the yield are discussed. For a number of initial disturbances in the input variables an optimal compensation has been determined from the following NLP problem:

$$\max_{\delta u} \{\delta x^T \mathbf{b} + \delta x^T B \delta x\} \quad \|\delta x\|_2 \leq 2 \tag{10}$$

where $\delta x = (d_1,\ldots,d_m,\delta u_1,\ldots,\delta u_{k-m})^T$ (compare with Eq. 2). Note that for $m = 0$ the optimal process operation conditions, as a result of a formal optimization, will be found again.

Table 4. Final Simplex

Vertex		1	2	3	4	5	
Reaction Temperature	ξ_1	64.1	64.5	64.3	64.2	64.3	(°C)
Reaction Time	ξ_2	2.29	2.22	2.33	2.33	2.22	(h)
Amount of KOH	ξ_3	545	552	537	541	553	(g)
Amount of Formaldehyde	ξ_4	438	463	454	449	465	(g)

For the first run (see Table 5) the reaction temperature has been limited to 63°C (that is, $\delta x_1 = -1$). This process disturbance was compensated by adjusting the other input variables according to Eq. 10, where β and $\mathbf{B}$ have been estimated from the preceding experiments. It appeared that the reaction time had to be extended with respect to the nominal conditions with 0.44 h, the total amount of KOH to be charged was 572 g instead of 500, and the amount of formaldehyde was 405 g instead of 425. The predicted output associated with these adjustments was 81.4%, while the actual yield was only 74.6%. For the third run this deviation was even larger due to the sudden occurrence of a side-reaction which spoiled the batch. Also, for runs five and seven rather large errors appeared. Summarizing, at first sight the results of this control strategy are rather disappointing. However, we must keep in mind that, in fact, we should have rejected the fitted model (Eq. 7); the value of F_L (Table 2) is larger than the upper 1% point of the associated F distribution. So, better results might have been obtained when a more adequate model was used, or when the disturbance were chosen smaller. It should be noted that application of the model without the nonsignificant terms would not substantially deviate from the implemented strategy, at least for the first runs.

Conclusions

The application of flexible recipes can be a powerful tool in the (batch) process industries for increasing the productivity. Both a model-based and an empirical optimization procedure have been performed to a specific industrial application: the batch-wise production of benzylalcohol on a pilot-plant scale. It appears that from these procedures, response surface methodology and (Nelder-Mead) simplex EVOP, different solutions are found, which is caused by the special form of the response surface (mini-max). Therefore, we conclude that for more complex response surfaces RSM is preferable to simplex EVOP. In addition to the actual information about the process, the graphically presented evaluation of, for example, measured

Table 5. Experiment Results of Feedforward Control

Temperature of Reaction $\xi 1$	Time of Reaction $\xi 2$	Amount of KOH $\xi 3$	Amount of Formaldehyde $\xi 4$	Predicted Effect y	Measured Effect z	Prediction Error e
63.0*	1.94	572	405	81.4	74.6	−6.8
65.2	**1.04**	446	459	73.7	76.5	2.8
65.1	2.05	**450**	467	82.4	47.0	−35.4
63.9	2.08	565	**375**	80.3	76.8	−3.5
63.0	**1.00**	430	437	55.6	39.9	−15.7
65.3	**1.00**	**450**	446	70.0	77.7	7.7
65.3	**1.00**	468	**375**	63.7	50.9	−12.8
63.0	2.15	**450**	397	63.7	63.2	−0.5
63.2	2.10	**450**	**375**	64.0	63.9	−0.1

*Bold-faced numbers represent the imposed disturbance.

process output, parameter estimates or residuals is also of great importance. The proposed adaptive feedforward control strategy offered rather large prediction errors, which can be attributed to the inadequacy of the identified second-order model.

Acknowledgment

This research was supported by the Netherlands Technology Foundation (STW), Grant No. TST88.1610. I am indebted to Eddy van Selow who performed a large number of the experiments and to Prof. John Rijnsdorp for his comments on a first draft of the article.

Notation

b = vector of first-order parameter estimates
B = matrix of second-order parameter estimates
d = disturbance vector
e = output error
k = number of independent inputs
m = number of disturbances
M = matrix of eigenvectors
N = number of experiments
q = white noise vector
u = control input vector
x = (coded) input vector
y = model response
z = measured process output

Greek letters

β = vector of first-order parameters
β_0 = nominal model response
$\mathbf{B}$ = matrix of second-order parameters
δ = difference operator
ϵ = guaranteed (output) error
θ = vector of transformed first-order parameters
Λ = diagonal matrix of eigenvalues
ν = vector of input bounds
ξ = vector of nominal inputs

Literature Cited

Box, G. E. P., "Evolutionary Operation: A Method for Increasing Industrial Productivity," *Appl. Statist.*, **6**, 3 (1957).

Box, G. E. P., and N. R. Draper, *Evolutionary Operation*, John Wiley, New York (1969).

Dem'yanov, V. F., and V. N. Malozemov, *Introduction to Minimax*, Halsted Press Book, John Wiley, New York (1974).

Grossman, I. E., K. P. Halemane, and R. E. Swaney, "Optimization Strategies for Flexible Chemical Processes," *Comput. Chem. Eng.*, **7**(4), 439 (1983).

Grossman, I. E., and C. A. Floudas, "Active Constraint Strategy for Flexible Analysis in Chemical Processes," *Comput. Chem. Eng.*, **11**(6), 675 (1987).

Keesman, K. J., "Membership-set Estimation Using Random Scanning and Principal Component Analysis," *Math. Comp. Simul.*, **32**(5,6), 535 (1990).

Keesman, K. J., "Use of Programming Methods for Flexible Recipes in Batch Process Control," *Proc. IFAC Symp. Advanced Chemical Control*, Toulouse, pp. 355 (October 1991).

Keesman, K. J., and G. van Straten, "Identification and Prediction Propagation of Uncertainty in Models with Bounded Noise," *Int. J. Control*, **49**(6), 2259 (1989).

Milanese, M., and R. Tempo, "Optimal Algorithms Theory for Robust Estimation and Prediction," *IEEE Trans. Autom. Control*, Vol. AC-**30**(8), 730 (1985).

Nelder, J. A., and R. Mead, "A Simplex Method for Function Minimisation," *Comput. J.*, **7**, 308 (1965).

Rijnsdorp, J. E., *Integrated Process Control and Automation*, Elsevier Sci. Publ., Amsterdam (1991).

Spendley, W., G. R. Hext, and F. R. Himsworth, "Sequential Application of Simplex Designs in Optimisation and EVOP," *Technometrics*, **4**, 441 (1962).

Tempo, R., B. R. Barmish, and J. Trujillo, "Robust Estimation and Prediction Using ARMA Models: A Nonstatistical Approach," *IFAC Symp. Identification and System Parameter Estimation*, Beijing, pp. 821 (July 1988).

Verwater, Z., Internal Report, NR-1641, Technical University of Eindhoven, p. 16 (in Dutch) (1990).

Walter, E., and H. Piet-Lahanier, "Estimation of Parameter Bounds from Bounded-Error Data: A Survey," *Math. Comp. Simul.*, **32**(5,6), 449 (1990).

Young, P. C., *Recursive Estimation and Time Series Analysis*, Springer-Verlag, Berlin (1984).

Manuscript received March 25, 1992, and revision received Sept. 11, 1992.